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## CREEP–RUPTURE CHARACTERISTICS OF POLYCRYSTALLINE OXIDE CERAMIC TO 1600°C

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Creep in polycrystalline oxide ceramics at temperatures 1400 – 1600°C occurs by diffusion. The creep curves are very sensitive to the presence of nonequilibrium defects and stress concentrators which were “annealed” during cooling after high-temperature kilning of the materials. Apparently, rupture is due to vacancies forming in stretch-zone sections tied to crystal boundaries followed by their coagulation on the boundaries. Cracks form and develop differently in different materials, depending on the crystal-chemical properties of the materials.

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**Key words:** polycrystalline oxide ceramic, creep, rupture.

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Polycrystalline ceramics under a mechanical load at temperatures to 1200 – 1300°C behave as a brittle material and rupture after a small elastic deformation. In tests performed at high temperatures some plastic deformation, though weak, is observed. Such behavior is due to the particular structural complexity of a ceramic at the crystal-lattice and the micro- and macrostructural levels; it is determined by the ionic-covalent type chemical bond in ionic compounds [1].

The mobility of the lattice elements is low because of the high strength and directionality of the anion – cation bonds. In addition, two or more types of ions differing by their chemical nature, radii, and charge form unit cells of substantial size comprised of two or more sublattices. All this also slows the movement of ions through the lattice. The structure of the ceramics is complicated. Aside from special cases these are polycrystalline solids consisting of several phases, including a glass phase and pores. Such a structure intensifies inertness and the resistance inhering in the solid state to external actions, in the case of ceramics right up to pre-melting temperatures [2].

It is well known that deformation of a solid is the result of two processes — slip along crystallographic planes (shear,

rotation) and plastic or viscous flow. In polycrystalline ceramic materials, the structural complexity of the unit cells impedes slip processes and the diffusion of point defects effectuates plastic deformation. Likewise, because friction forces in the lattice are strong the observed motion of dislocations, boundaries, and so forth proceeds vacancy-by-vacancy. When a continuously distributed glass or liquid phase is present viscous flow is observed in refractories with complex composition [3].

The study of high-temperature creep (above 1600°C) has established that the ordinary creep strain versus time curves are straight lines [4]. Exceptions have been observed only in some special cases where the structure of the material changes gradually as temperature increases. At the same time tests performed on metals and alloys show the presence of three stages of creep: the primary (transient) stage followed by a secondary stage of steady-state creep and tertiary stage of accelerated creep prior to rupture [5].

The objective of the present work is to study the creep characteristics of polycrystalline oxide ceramic under a mechanical load in the temperature range where a transition from brittle to plastic fracture occurs. The tests were performed on materials based on periclase, alumina magnesia spinel, and corundum — materials often used in high-temperature equipment. Each type of ceramic was represented by two modifications differing by the predominant size of the crystals (according to optical microscopy data): periclase —

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**TABLE 1.** Properties of the Experimental Samples

Mix index	Ceramic type	Predominant crystal size, $\mu\text{m}$	Apparent density, $\text{g/cm}^3$	True porosity, %	Bending strength, MPa, at temperature, $^{\circ}\text{C}$				
					20	1400	1450	1500	1550
K-15	$\text{Al}_2\text{O}_3$	15	3.90	2.5	150	70	56	52	50
K-35	$\text{Al}_2\text{O}_3$	35	3.90	2.5	125	60	54	51	42
ShP-10	$\text{MgAl}_2\text{O}_4$	10	3.49	2.5	150	35	22	18	15
ShP-25	$\text{MgAl}_2\text{O}_4$	25	3.50	2.2	120	30	20	15	13
P-12	$\text{MgO}$	12	3.47	3.0	120	65	52	45	40
P-25	$\text{MgO}$	25	3.50	2.2	105	60	73	62	55

12, 25  $\mu\text{m}$ ; spinel — 10, 25  $\mu\text{m}$ ; and, corundum — 12, 25  $\mu\text{m}$ . The properties of the samples are presented in Table 1.

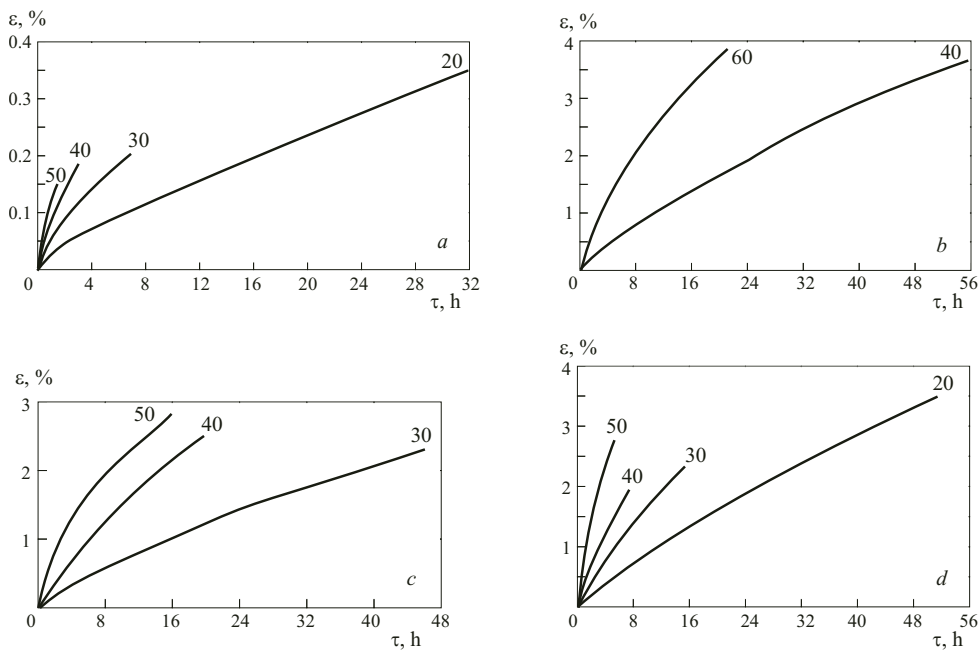
The experiments were performed at temperatures 1400 – 1550 $^{\circ}\text{C}$  under loads to 60 MPa, close to the ultimate strength of the materials. Compared with the results of creep studies [4] the data obtained give a more complete picture of the behavior of dense polycrystalline ceramics. It is of special interest to establish a connection between the characteristics of creep, specifically, the rate at the steady-state stage and the long-term strength, i.e., the time and creep strain to rupture. At the same time the objects chosen make it possible to determine the effect of their crystallographic characteristics on the course and mechanisms of the process — periclase and spinel crystallize into a comparatively simple cubic lattice while corundum crystallizes into a complex hexagonal lattice. Even though the difference of the melting temperatures is substantial (periclase — 3100 K, corundum — 2320 K, spinel — 2400 K), the creep rates of the materials do not differ as much. The lowest creep rates are observed for corundum, followed by periclase and spinel [1].

### CREEP STRAIN CURVES

Curves of the creep strain to rupture were obtained in the tests. In general, these curves included all three stages of creep — transient, steady-state, and accelerated. Figure 1 displays typical measurements of the creep strain of the samples. Periclase ceramic with predominant crystal size 25  $\mu\text{m}$  (P-25 in Table 1) is chosen as an example. It is evident that rupture almost always starts during the steady-state creep stage.

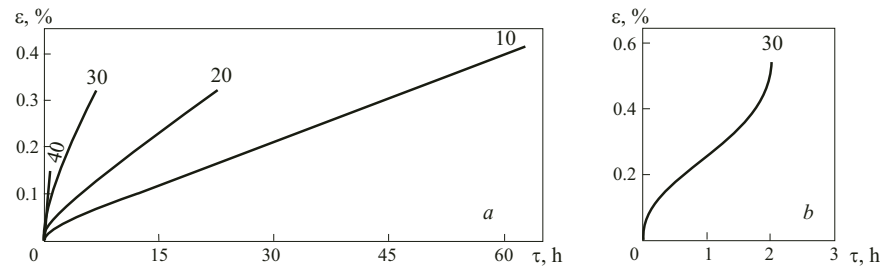
First, a behavioral characteristic common to all experimental materials, specifically, the individual character of the deformation processes in each specific sample, should be noted. Irrespective of the test temperature and load curves all samples show steady-state creep, but the transient and accelerated creep stages could be absent. The last stage before rupture was observed only in individual cases, while the first stage was observed much more often — in more than 50% of the tests.

In the opinion of the authors, such unstable behavior of the samples is due to the presence in the samples of stress



**Fig. 1.** Creep strain  $\epsilon$  curves for P-25 periclase samples (Table 1) at temperatures 1400 (a), 1450 (b), 1500 (c), and 1550 $^{\circ}\text{C}$  (d) (the load, MPa, is indicated on the curves).

**Fig. 2.** Creep strain curves for K-35 corundum samples (see Table 1) at temperatures 1500°C (*a*) and 1550°C (*b*) (the load in MPa is indicated on the curves).



concentrators and nonequilibrium defects, which were partially “annealed” during cooling after kilning at high temperature. Indeed, at high test temperatures the stresses at concentrators and defects relax during heating. However, as temperature decreases, the relaxation rate decreases and the concentrators can be annealed. Since the samples were not annealed after kilning, their behavior in subsequent creep tests will depend randomly on the conditions under which they were found during cooling under the complex conditions of a flame furnace.

The form of the curves in Fig. 1 is understandable from the standpoint considered here. The typical curves with two sections — transient and steady-state creep — are obtained at the test temperature 1400°C. As the load increases, the creep rate increases, the time of the secondary stage decreases, and the times to rupture and the deformation of the sample decrease. In all cases rupture starts during steady-state creep. However, the picture is somewhat different at temperatures 1450 and 1500°C. Practically steady-state creep is observed under relatively light loads (30–40 MPa), while transient creep appears with increasing load. The time to rupture of the sample decreases and the deformation of the sample increases somewhat. The sample tested at 1550°C and 30 MPa showed accelerated creep prior to rupture. Once again, typical curves with two creep sections are obtained at 1550°C. However, the magnitude of the deformation up to rupture first decreases and then increases with increasing load.

Similar data were obtained for two other types of ceramic. As an example, the creep strain curves for corundum ceramic with predominant crystal size 35  $\mu\text{m}$  are presented in Fig. 2. The first two sections of the creep strain curve are observed at temperature 1500°C. As the load increases, the time of the transient period changes nonmonotonically; rupture starts during steady-state creep. However, at 1550°C the accelerated creep stage was observed before rupture.

Thus, for the experimental testing conditions (1400–1500°C, loads to 60 MPa) the flow of the creep and rupture processes is different in each fully sintered, polycrystalline, ceramic sample. This is different from the behavior of such materials at higher temperatures (above 1600°C) and small loads (to 3 MPa) for which short-time creep tests have been performed [4]. In the latter case only linear time dependences of the creep strain, i.e., steady-state creep, were observed. The foremost reason for this is the high test temperature,

heating to which causes the stresses in the initial samples to relax when creep occurs by diffusion.

Experiments on thermal aging have shown [6] that practically no increase of the crystal size is observed for oxide-based polycrystalline materials with soaking times to 2000–3000 h at 1300°C. The periclase samples are exceptions. They show small crystal growth under these conditions. It is well known that larger crystals are obtained by means of diffusion. Thus it can be supposed that diffusion in the experimental materials is slow at temperatures to 1300°C. The interval 1300–1600°C is a transient interval where as temperature increases during creep the diffusion processes predominate. Brittle rupture occurs at lower temperatures.

Under the indicated conditions such regions were also recorded in experiments performed to determine the creep mechanisms [4]. On transition from high temperatures and small loads to lower temperatures but comparatively high loads the creep rate decreases considerably as result of a decrease of the diffusion mobility of the atoms. Boundary diffusion mechanisms operate in this region, but the plasticity associated with them is low. In [7] it is shown that under mechanical loads polycrystalline ceramics behave as brittle materials at temperatures to approximately 1200–1300°C. Diffusion-viscous flow mechanisms are activated at higher temperatures according to their energy energies — first boundary and then volume diffusion processes are activated. In the region of brittle behavior of the ceramic, because stress concentration occurs at the joints between crystals and near pores rupture occurs with practically with no flow strain.

The appearance of a transient stage during diffusion creep has also been observed in experiments on metals [8]. Researchers attribute this phenomenon to the presence of different types of nonequilibrium defects, which contribute to the total creep strain of the materials undergoing “annealing” in the course of the tests. Such defects are stress concentrators and arise when a ceramic is cooled from the high temperatures at which it is produced by conventional methods. The presence of concentrators is the reason why there are some differences in the behavior of the samples undergoing creep strain and rupture.

## MICROSCOPIC PICTURE OF RUPTURE

The microstructure of the materials after testing for long-term strength was investigated on polished sections in



**Fig. 3.** General picture of the macrostructure of ceramic samples after tests for long-term strength: *A*) compression zone; *B*) neutral zone; *C*) tension zone; *a*) shear region; *b*) region of porosity propagation.

reflected light. The part of the lateral surface of the samples that was subject to pure bending was studied. The general picture of the macrostructure of a sample is shown schematically in Fig. 3. A large number of pores, cavities, and cracks are present in the tension zone *C* (region *b*). A region *a* where surface shearing is sometimes observed exists in the compression zone *A*. The neutral zone *B* is completely free of pores and different kinds of cavities; the structure of this zone actually remains unchanged after the tests.

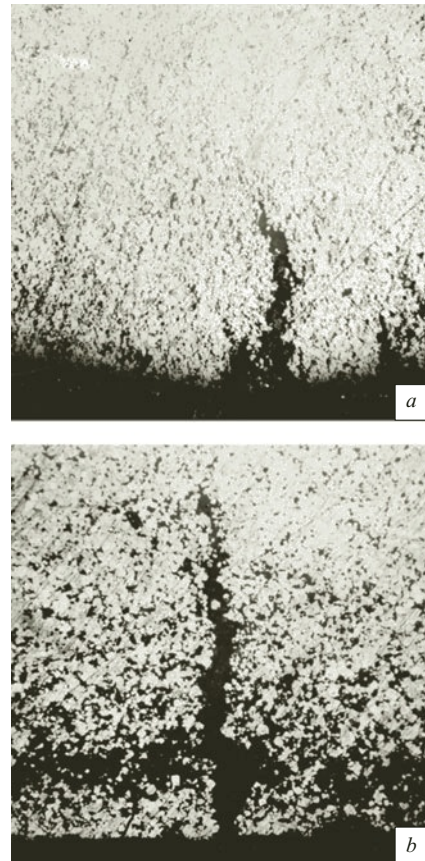
It was found that the character of porosity and microcrack formation is approximately the same for each type of ceramic studied, irrespective of the crystal sizes. The form of the microstructure of fine- and coarse-crystalline samples of periclase is presented in Fig. 4 as an example.

Because the tests established that in almost all cases rupture of the samples was observed without any appreciable increase of the creep rate it can be assumed that high porosity and microcracks form in a stretched sample at the steady-state creep stage, where cracks develop and rupture occurs along the most dangerous cracks. Judging from the published data [5], cracks and voids are not found immediately upon loading, i.e., at the primary creep stage. They appear after a certain critical creep strain.

The formation of pores and microcracks is due to tensile stresses. Pores coalesce in the course of plastic deformation, which subsequently results in the appearance of microcracks. In the process, as a rule, several cracks penetrating into the interior volume of the sample exist along the entire length of the tension zone; these cracks appear at sites on the surface where the probability of finding microdefects is highest. Depending on the plastic properties of the ceramic the samples are distinguished by the degree of the porosity. The highest porosity (near a crack) is observed in periclase samples in which high flow strain is also observed.

Pores coalesce into cavities, which, joining with one another, form cracks of considerable size. In all periclase samples, the maximum length of post-test cracks reaches one-half the height of the sample (Fig. 5) and is larger than the cracks in spinel and corundum samples (Figs. 6 and 7).

No visible cracks were observed in corundum (Fig. 7). Creep strain is lowest in corundum, and the character of crack propagation in corundum differs from that in periclase. Just as in corundum, no visible voids were found in spinel (see Fig. 6). Cracks develop by coalescence of microcavities



**Fig. 4.** Exterior view of the porosity and microcracks: *a*, *b*) periclase P-12 and P-25 according to Table 1 ( $\times 40$ ).

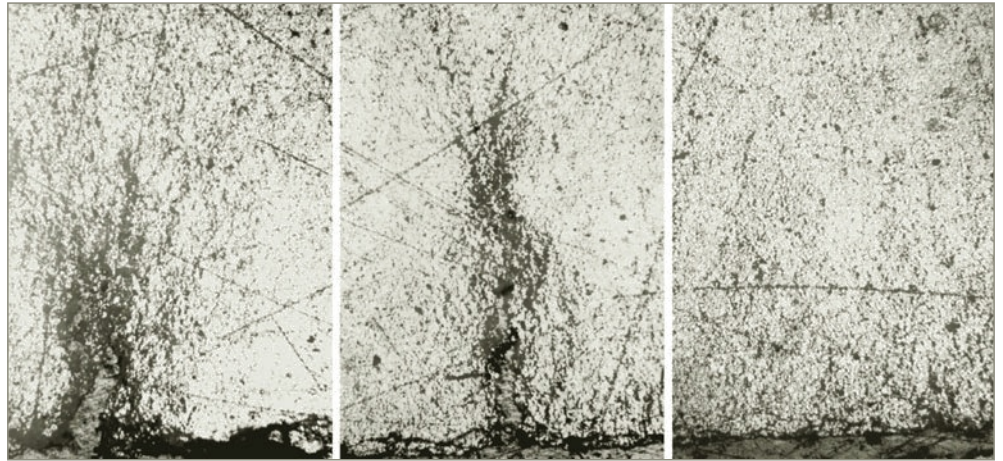
without the formation of substantial porosity. Plastic deformation of the tension zone “loosens” the crystal boundaries, one result of which is substantial pitting of the particles even when samples are etched during the preparation of polished sections (see Fig. 5).

The formation, coalescence, and propagation of microcracks along the weakest sites, i.e., along the boundaries between crystals, occur in all three materials. Crack development in them during long-term strength tests is shown in Figs. 5–7.

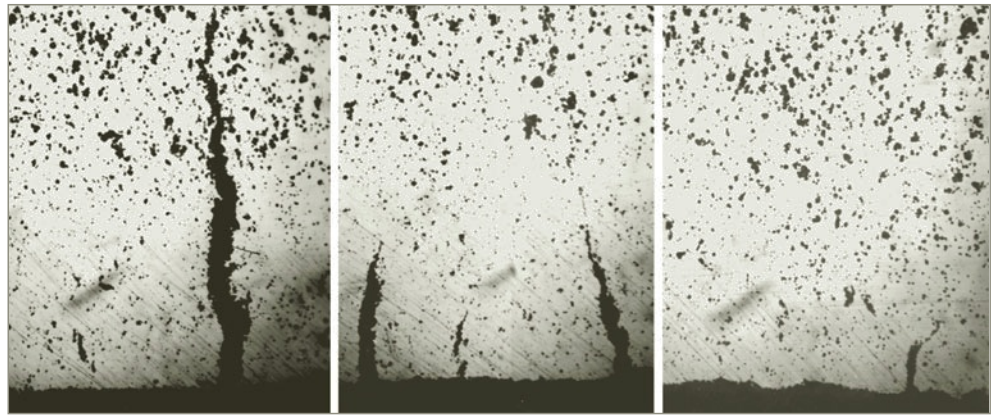
Compared with the starting crystals no substantial changes of the crystal sizes and shapes were found over the entire cross section of a sample, i.e., over all three zones — compressed, neutral, and stretched. An example of the microstructure of the neutral and compressed zones of a periclase sample after testing for long-term strength is presented in Fig. 8. Probably, the temperatures at which the studies were performed are too low for any appreciable crystal growth to occur, i.e., the diffusion mobility of the atoms (ions) is low. Here lies the difference in the behavior of the samples at temperatures to 1600°C from the results obtained at higher temperatures [4].

A change in the character of the rupture of the materials as a function of temperature and stress was not observed in

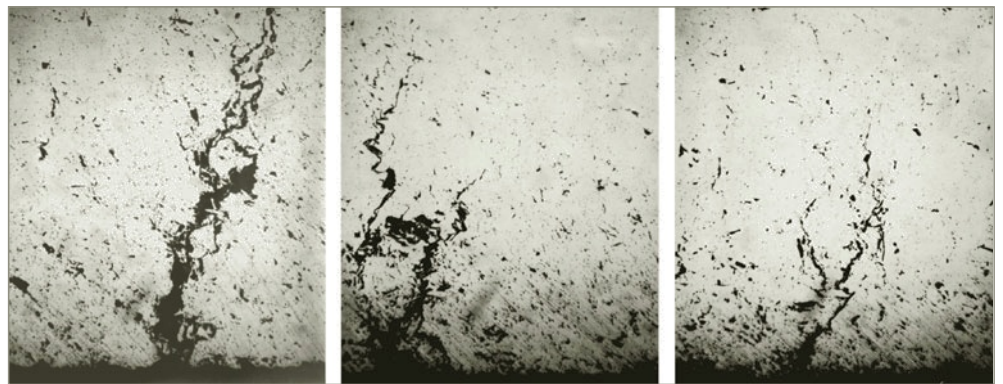




**Fig. 5.** Typical cracks and pores in the tension zone of periclase samples ( $\times 40$ ).



**Fig. 6.** Typical cracks and pores in the tension zone of spinel samples ( $\times 40$ ).



**Fig. 7.** Typical cracks and pores in the tension zone of corundum samples ( $\times 40$ ).

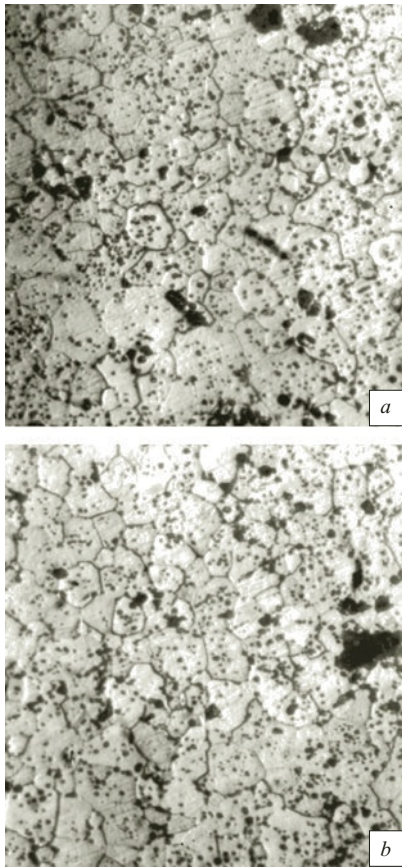
all cases. The shears observed in the compression zones of the ruptured samples (see Fig. 3, region *a*) are due to shearing forces arising during bending.

Judging from the creep strain curves, periclase ceramic, just as the coarse- and fine-crystalline ceramic, possesses comparatively high plasticity under the test conditions adopted. Thus, the deflection prior to rupture at different temperatures and stresses lies in the range 1.5 – 3.6% with the exception of coarse-crystalline samples which at 1400°C showed a 10 times lower value. The deflection occurring

prior to rupture and the time to rupture, as a rule, decrease with increasing temperature and stress.

Likewise, spinel ceramic is characterized by quite high plasticity under the test conditions. The two experimental forms of this ceramic have similar creep strain curves. The deflections prior to rupture are 0.9 – 1.0% and any difference is negligible; at 1450 and 1500°C the coarse-crystal samples show a large deflection prior to rupture — 1.4 – 2.3%.

Corundum ceramic has low plasticity under the test conditions. Even at 1550°C and 20 MPa the deflection before



**Fig. 8.** Microstructure of periclase (P-12 in Table 1) after testing for long-term strength ( $\times 340$ ): *a*) neutral zone; *b*) compressed zone.

rupture was only 0.55%. Compared with coarse-grain samples fine-grain samples show a higher creep rate, larger deflection before rupture, and shorter time to rupture.

In summary, a study of creep strain and long-term strength established that for the test conditions adopted cracks form and develop differently, depending on the crystal-chemical particulars of the materials.

The ionic component of the chemical bond predominates in the simple cubic lattice of periclase. For this reason the diffusion mobility of the atoms is high, and rupture is due to the coagulation of a substantial number of vacancies on the boundaries of the crystals in the tension zone with appreciable irreversible deformation. Vacancies are formed throughout the entire volume of the tension zone, but their concentration is higher near crystal boundaries.

The covalent component of the chemical bond predominates in the complex hexagonal lattice of corundum. For this reason, the diffusion mobility of the atoms is much lower than in the periclase lattice, even though the melting temperature of corundum is appreciably lower. Correspondingly, the flow strain of corundum is also appreciably smaller under the conditions considered. Most likely, rupture occurs as a result of the appearance of vacancies in the tension zone which are confined to the crystal boundaries as well as the coagulation of vacancies on the crystal boundaries.

In the spinel lattice, which is of intermediate complexity but closer to the corundum lattice, the rupture mechanism during creep is closer to that of corundum, judging from petrographic data.

## CONCLUSIONS

1. Under a mechanical load in the temperature interval 1400 – 1600°C the formation, consolidation, and propagation of microcracks are observed along the weakest locations, specifically, the intercrystalline sections.
2. At temperatures 1400 – 1600°C and loads to 60 MPa the flow of these processes depends on the particular strain properties of the materials, specifically, the type of chemical bond.
3. No substantial changes of the crystal sizes of the samples were observed after the tests.

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